

**EFFECTS OF POLYETHYLENE TEREPHTHALATE (PET)  
FROM PLASTICS WASTE ON THE THERMAL PROPERTIES OF ASPHALT**

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**INTRODUCTION**

Asphalt concretes are widely used for pavements. Asphalt itself is a bitumen and although it occurs naturally, it is mostly obtained as a byproduct of oil production. However, its thermoplastic characteristics cause difficulties with roads. It has been demonstrated that the apparent glass transition temperature of asphalt is near zero degree celsius [1,2]. Thus, asphalt concretes are susceptible to low temperature cracking that may lead to fracture. Further, in high summer temperatures asphalt undergoes flow or creep. The stability of asphalt paving surfaces requires that it does not flow or creep under heavy load. Numerous investigations are underway involving the modification of asphalt with different fillers that may improve its pavement performance. For example, the modification of asphalt with polymers leads to bituminous materials having improved properties and allows the utilization of waste products [3].

In this investigation, the effect of polyethylene terephthalate (PET) obtained from used plastic bottles on the thermal properties of asphalt, specifically its effect on the thermal expansion coefficients, glass transition temperatures, melting points, and flow properties of asphalt, PET, and asphalt/PET mixture samples, is evaluated. Polyethylenes have been confirmed as potentially useful modifiers for increasing the low temperature fracture toughness of the asphalt and may also contribute to pavement stability at elevated temperatures by minimizing distortion due to creep [4]. The abundance of used plastic bottles that contributes to the litter problem and the resultant environmental pollution caused by conventional non-degradable plastics makes PET fillers economically attractive for improving the performance of asphalt concrete paving materials.

**EXPERIMENTAL TECHNIQUES**

**Sample Preparation**

Asphalt of grade AC-20 certified to comply with the state of New York highway specifications was provided by Prima Asphalt Conc., Inc. Polyethylene terephthalate (PET) filling material was obtained from used plastic soda bottles that were first cut into

strips about 1/4 inch in thickness and then ground using a grinder with a 20 mesh sieve.

The asphalt/PET mixtures were prepared by slowly adding a weighted amount of PET to a known quantity of asphalt maintained at 120°C while the mixture was vigorously stirred. This temperature is below the melting point of PET but above that of asphalt. After several minutes of stirring, the mixtures were left to cool to room temperature, and stored until used for testing. Sample concentration ranged from 10% to 50% PET by weight.

#### **DSC Measurements**

The glass transition temperatures ( $T_g$ ) and the melting points of asphalt, PET, and asphalt/PET mixtures were measured using differential scanning calorimetry (DSC). The samples were heated at a 10 °K/min rate. between 100°K and 600°K.

#### **TMA measurements**

Thermomechanical analysis (TMA) was employed to determine the expansion coefficients, glass transition temperatures ( $T_g$ ), melting points, and flow properties of asphalt, PET, and asphalt/PET mixture samples. The temperature range of the measurements was between 100°K and 600°K. Cylindrical samples approximately 9.5 millimeter in diameter and between 0.3 and 3.0 millimeter in thickness were heated at a rate of 5°K/min. Expansion coefficients were calculated from the TMA expansion profiles using aluminum expansion coefficient data to determine the dynamic calibration constant. A 5 grams mass supplied the compressive force to the samples. Flow properties were measured using parallel plate rheometry. Samples were compressed by a 50 gram load.

### **DISCUSSION AND RESULTS**

#### **Differential Scanning Calorimetry**

Based on several DSC runs, the average  $T_g$  for pure asphalt was calculated to be -41.4°C. Values for asphalt  $T_g$ 's in the literature range between -0.4°C to -53°C [5,6]. In general, the change in the heat flow curve slope indicating the glass transition temperature was very small and sometimes even undetectable; consequently, it was very difficult to characterize the samples using DSC technique. However, in the cases where it was detected, the DSC results were reproducible and yield  $T_g$  values within the 5% accuracy of the instrument.

The PET/asphalt mixture DSC measurements yield  $T_g$  values in the same range as those obtained for pure asphalt. There was no

indication that the presence of PET fillers affected the Tg of asphalt. Fig. 1 is the plot of the Tg versus PET concentration.

DSC was also used to measure the melting point (Tm) of PET. For pure PET the average melting point was determined to be 235.4°C. As with the asphalt Tg, there was no indication that the melting point of PET was affected by the presence of asphalt. The average melting temperature of PET calculated using results obtained from pure PET and PET/asphalt mixtures was estimated as 234.9°C. Fig. 2 is a plot of the PET Tm values versus mixture composition. Literature gives a range for the melting point of pure PET between 250°C and 265°C. Taking into account the difference in techniques used, the accuracy of the instrumentation, and the fact that the DSC results were obtained using PET from used plastic bottles instead of pure PET, The discrepancy between the DSC findings and literature values is not unexpected.

#### Thermomechanical Analysis

As stated previously, the average Tg value for asphalt was estimated at -41.4°C. In general, the asphalt Tg obtained using the TMA technique is about 70 degrees higher than those obtained by the DSC technique. This discrepancy may be attributed to the fact that these techniques measure different phenomena. The Tg by DSC corresponds to a change in the ability of the sample to absorb heat, while the TMA detects the Tg as a change in the thermal expansion profile of the sample.

The average Tg for asphalt as determined by the TMA technique was estimated as 39°C; the value estimated including glass transition temperatures obtained for the PET/asphalt mixtures is 33.4°C. A plot of the average asphalt Tg values versus PET concentration is shown in Fig. 3.

The melting point of PET was also measured using the TMA technique. These measurements yield 248.3°C as an average Tm for PET, and an average Tm value of 243.8°C for the asphalt/PET mixtures. These TMA results are about 10 degrees higher compared to the DSC findings and that much closer to the literature values. In Fig 4, the average Tm values are plotted as a function of asphalt concentration.

Examination of the TMA curve shows that PET exhibits a glass transition temperature at about 81°C that was not detected when the DSC technique was used. PET TMA measurements yield an average Tg of 81.8°C with values ranging from 80.4°C to 85.8°C. The literature [7] value for PET Tg is 81°C. Unlike the previous findings, the glass transition temperature of PET does seem to be affected by the presence of asphalt; so much that it was not possible to detect it with certainty from examination of the TMA curves for PET/asphalt mixtures. Furthermore, when the PET samples were subjected to a heating cycle, the Tg could not be detected.

The expansion coefficients were determined from TMA expansion/contraction profiles. An example of a TMA expansion/contraction profile for a PET sample is shown in Fig 5. Note that in this figure the slope of the TMA curve noticeably changes after the crossing of the glass transition temperature. For PET samples, two distinct regions are observed: a region that ranges from liquid nitrogen temperatures to about 81°C, the glass transition temperature of PET, and second one that extends from 81°C to the melting temperature. The average expansion coefficient for the first region is 109  $\mu\text{m}/\text{m}/^\circ\text{C}$ . The average expansion coefficient for the second region was estimated to be 955  $\mu\text{m}/\text{m}/^\circ\text{C}$ . The expansion coefficient for asphalt was found to be 290  $\mu\text{m}/\text{m}/^\circ\text{C}$ .

The addition of PET to asphalt has a more pronounced effect on the expansion coefficient. The addition of 10 wt% PET to asphalt reduces the expansion coefficient to 108  $\mu\text{m}/\text{m}/^\circ\text{C}$ . 20 wt% PET has an expansion coefficient of 89, 30 wt% PET has 85, 40% PET has 83, and a 50 wt% PET mixture has an expansion coefficient of 76  $\mu\text{m}/\text{m}/^\circ\text{C}$ , in the first region.

Additional information can be gained by studying the TMA derivative curve, using the parallel plate rheometer accessory [8]. The amplitude of the derivative curve will vary in intensity with the flow of the sample. The temperature of the peak (Ti) corresponds to the point of maximum flow rate. In general, the peak flow rate for PET/asphalt samples are very similar to those of PET. The Ti values range from 248.2°C for a 10/90 PET/asphalt sample to 252.3°C for a PET sample. This suggests that even for 10% PET concentration the overall flow of the mixture is dominated by the flow properties of PET. The PET filler acts as a binder restricting the flow of the asphalt until the filler itself starts melting. The inflection temperatures for pure asphalt range between 48.4°C to 58.6°C with an average Ti of 52.8°C. That is, the maximum flow rate for asphalt occurs at about this temperature.

Fig. 6 is a typical TMA plot for a 10/90 PET/asphalt sample featuring a derivative curve with two inflection points. The first one at 47°C corresponds to the maximum flow rate of asphalt. It is followed by a plateau due to the binding effects of the PET filler. The maximum flow rate of the mixture occurs at 252°C, the second inflection point. At this temperature, the PET filler is melting and flowing out of the rheometer plates together with the remaining asphalt.

A limited amount of viscosity information was also collected using the TMA parallel plate rheometer. The temperature dependent dimensional changes of a sample under a known load in the rheometer are converted to viscosity [9]. As expected there is a change of several orders of magnitude in the viscosity over a narrow temperature range as the sample starts to deform in the shear mode, due to thermal softening, see Figure 7. In the fluid region, the viscosity decreases with increasing temperature.

## CONCLUSIONS

- . Thermomechanical analysis and differential scanning calorimetry are useful techniques and yield reproducible results in obtaining melting points, glass transition temperatures, expansion coefficients, and the point of maximum flow rate (inflection temperature).
- . In general, asphalt's glass transition temperature does not seem to be affected by the presence of polyethylene terephthalate fillers.
- . The PET's glass transition temperature was clearly detected by the thermomechanical analysis technique even though the DSC measurements do not show a T<sub>g</sub> for PET.
- . The overall flow of the PET/asphalt mixtures is dominated by the flow properties of the PET.

## ACKNOWLEDGMENTS

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**Ave. Asphalt Tg Vs PET Concentration**  
Using DSC Technique

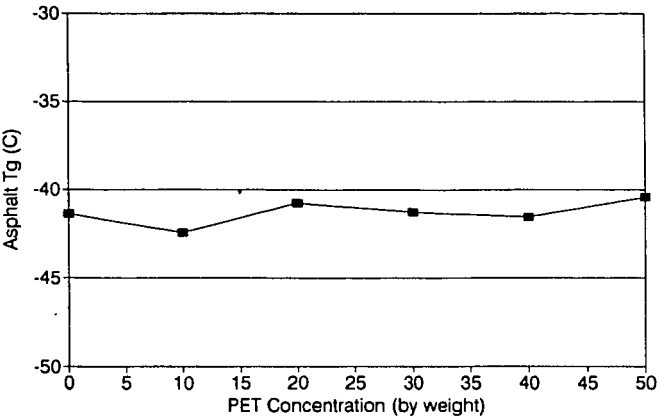


FIGURE 1.

**Ave. PET Tm Vs Asphalt Concentration**  
Using DSC Tehcnique

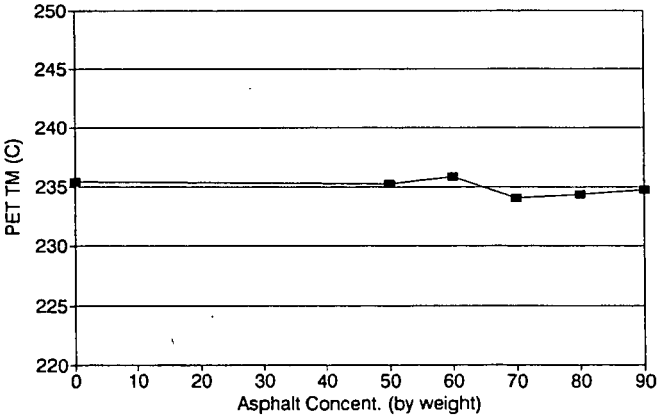


FIGURE 2.

### Ave. Asphalt Tg Vs PET Concetration Using TMA Technique

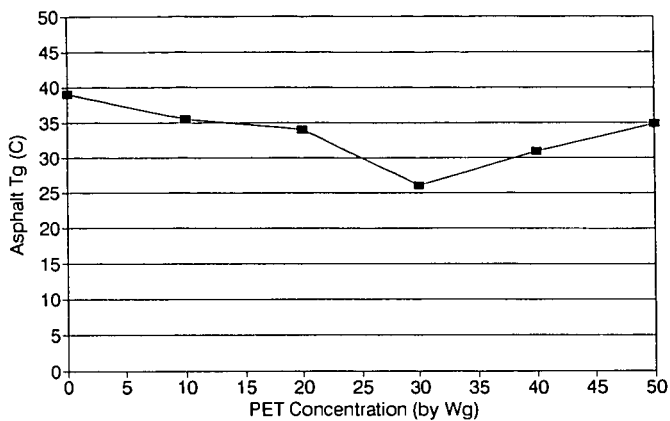


FIGURE 3.

### Ave PET Tm Vs Asphalt Concentration Using TMA Technique

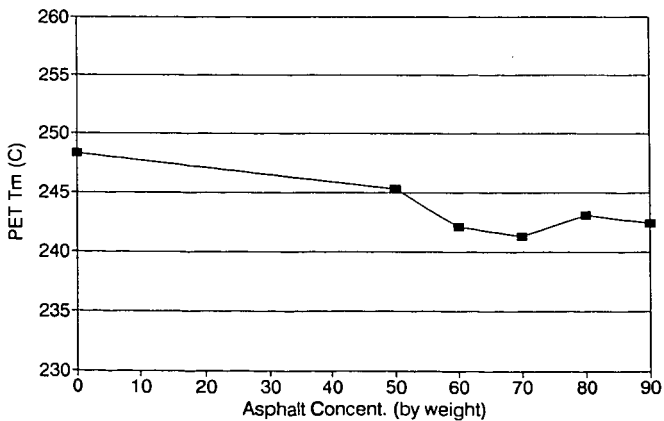


FIGURE 4.

Sample: PET (POWDER)  
Size: 2.2900 mm  
Method: TMA

File: C: PETASP-TMA.07  
Operator: RL  
Run Date: 15-Nov-90 11:57

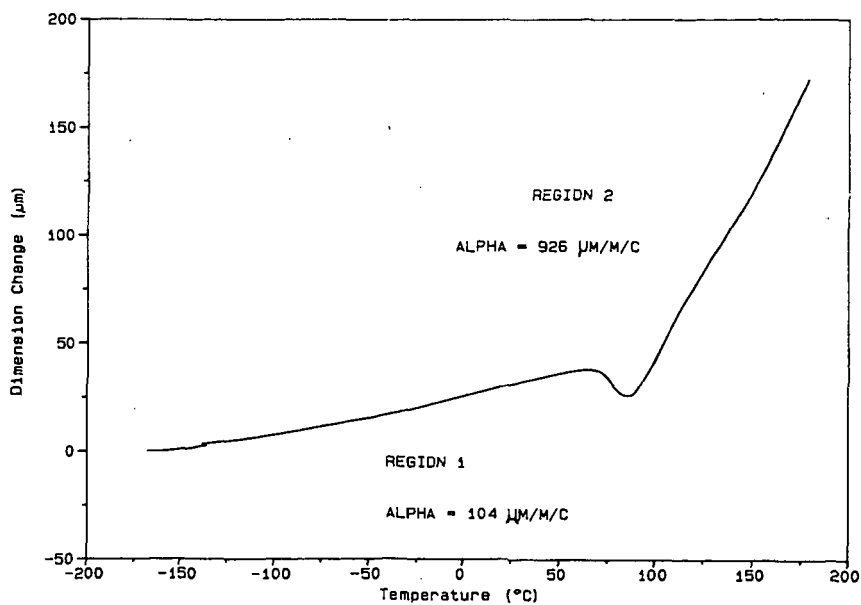


FIGURE 5.



Sample: PET/ASPHALT 10/90  
Size: 0.7746 mm  
Method: PPR

File: C: ASPE1.19  
Operator: RL  
Run Date: 23-Jan-91 14:28

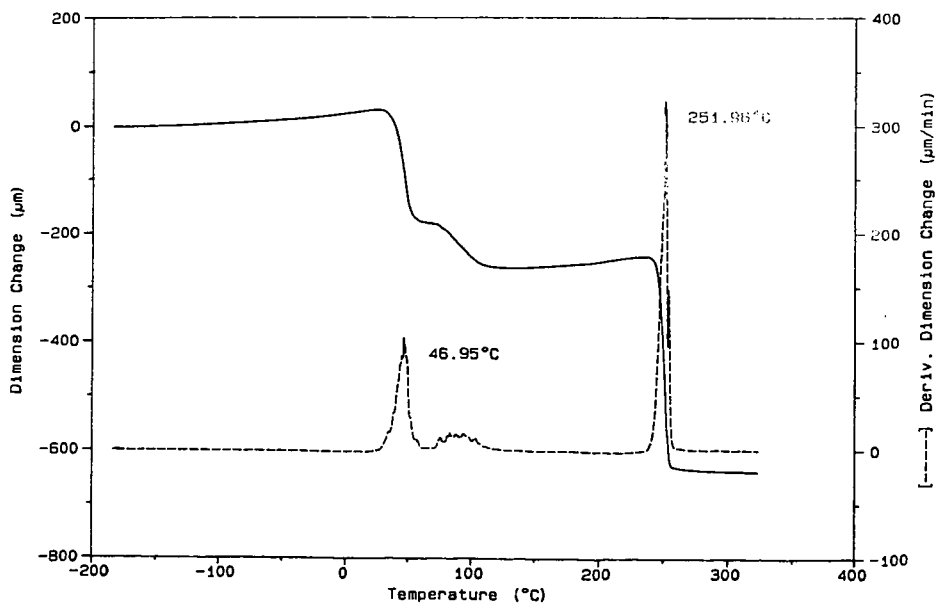


FIGURE 6.

# 40/60 PET/ASP. Visc. Vs Temp. (TMA PPR)

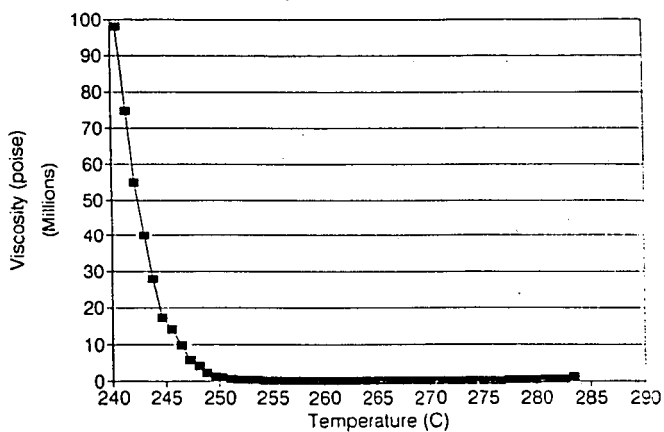


FIGURE 7.